

## Viscosity Prediction of Synthetic Lubricants from Temperature and Pressure Dependence of Dielectric Relaxation Time

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The dielectric permittance and the dielectric loss factor of several lubricating oils were measured at frequencies from 100 Hz to 1.5 MHz. The measurements were carried out under atmospheric pressure as a function of temperature and under fixed temperature as a function of pressure. Temperature and pressure dependence of dielectric relaxation time were investigated. The temperature dependence of relaxation time obeyed the Vogel-Fulcher-Tammann (VFT) law. We modified the VFT equation in order to express the dielectric relaxation time as a function of temperature and pressure. Furthermore, by taking into consideration the similarity of the temperature and pressure dependence between dielectric relaxation and mechanical relaxation, the prediction of high-pressure viscosity were conducted. The predicted results were compared with the viscosity data obtained from the falling-sphere type viscometer.

**Keywords :** Dielectric Relaxation, High Pressure, Viscosity, Synthetic Lubricants, Rheology

### 1. INTRODUCTION

Viscosity of lubricants is greatly dependent on temperature and pressure. Since the viscosity of lubricants increases exponentially with pressure, viscosity under elastohydro-dynamic condition increases by many order of magnitude. The viscosity measurement under high pressure is not so easy. For example, viscosity measurement using falling-body viscometer is time consuming. By contrast, dielectric properties can be measured accurately in a short time, and measurement under high pressure is relatively easy.

Dielectric relaxation is closely related to molecular mobility, and the similarity between dielectric relaxation and viscoelastic relaxation has been recognized. Accordingly, it is considered that the high-pressure viscosity of lubricants is predictable from the dielectric relaxation data. In this work, the dielectric measurements for several synthetic lubricants were performed under high pressure condition, and prediction of high-pressure viscosity was attempted.

### 2. EXPERIMENTAL

#### 2.1 Samples

The samples used in this work were di(2-ethylhexyl) phthalate (DOP), tricresyl phosphate (TCP), polyphenyl ether (5P4E) and CH<sub>3</sub> terminated poly(propylene glycol) having average molecular weight 1000 (PPGM1000). All samples were dried using molecular sieves.

#### 2.2 Dielectric measurements

Parallel capacitance  $C$  and conductance  $G$  of sample-filled capacitor were measured using an impedance analyzer in the frequency range from 100 Hz to 1.5 MHz. Dielectric constant  $\epsilon'$  and loss factor  $\epsilon''$  are calculated by the following equation:

$$\epsilon' = \frac{C}{C_0}, \quad \epsilon'' = \frac{G}{2\pi f C_0} \quad (1)$$

where  $C_0$  is air capacitance of the capacitor and  $f$  is frequency.

Measurements were carried out under atmospheric pressure as a function of temperature and under fixed temperature as a function of pressure up to 600 MPa. A two-terminal capacitor with an air capacitance of 78.2 pF was used for the measurements under atmospheric pressure, and under high pressure a coaxial cylindrical capacitor with an air capacitance of 142 pF was used.

### 3. RESULTS AND DISCUSSION

#### 3.1 Dielectric properties

Figure 1 shows the frequency dependence of the dielectric constant and the loss factor under different pressure at 293 K for TCP. Dielectric relaxation curves move to a lower frequency region with increasing pressure. Similarly, curves shift to a lower frequency region with decreasing temperature.

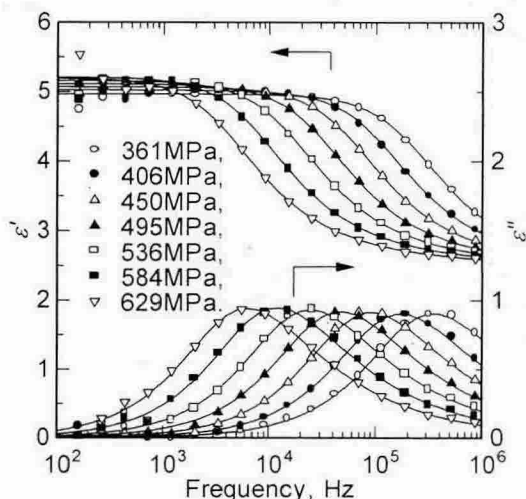


Fig. 1 Dielectric constant and loss factor for DOP as a function of frequency for given pressure at 293 K

The change in dielectric constant and loss factor against frequency can be described by HN (Havriliak-Negami) function [1] in the form:

$$\epsilon^*(\omega) = \epsilon_\infty + \frac{\epsilon_0 - \epsilon_\infty}{[1 + (j\omega\tau_{HN})^{1-\alpha}]^\beta} \quad (2)$$

where  $\epsilon^* = \epsilon' - j\epsilon''$  is the complex dielectric constant,  $\omega$  is angular frequency,  $\tau_{HN}$  is relaxation time and  $\alpha$  and  $\beta$  are fitting parameters ranging between 0 and 1.

### 3.2 Temperature and pressure dependence of dielectric relaxation time

Temperature dependence of dielectric relaxation time under atmospheric pressure obeyed VFT (Vogel-Fulcher-Tammann) relation of the form:

$$\ln \tau = \ln \tau_0 + \frac{DT_0}{T - T_0} \quad (3)$$

where  $\tau$  is relaxation time,  $T$  is temperature,  $\tau$ ,  $D$ , and  $T_0$  are fitting parameters. Yasutomi, Bear and Winer have derived the excellent viscosity-temperature-pressure equation by introducing the pressure dependence of the glass transition temperature. As is the case for the glass transition temperature, we assumed that the pressure dependence of  $T_0$  in equation (3) could be expressed by following function:

$$T_0(P) = T_0(P_0) + A_1 \ln[1 + A_2(P - P_0)] \quad (4)$$

where  $P$  is pressure,  $P_0 = 0.1$  MPa, and  $A_1$  and  $A_2$  are material dependent parameters. By substituting equation (4) into equation (3), we can obtain following dielectric relaxation time-temperature-pressure formula:

$$\ln \tau(T, P) = \ln \tau_0 + \frac{D[T_0(P_0) + A_1 \ln[1 + A_2(P - P_0)]]}{T - T_0(P_0) - A_1 \ln[1 + A_2(P - P_0)]} \quad (5)$$

Figure 2 shows the variation in dielectric relaxation time as a function of pressure for DOP. Solid lines show best-fit curves using equation (5). Temperature and pressure dependence of the dielectric relaxation time could be expressed sufficiently by equation (5).

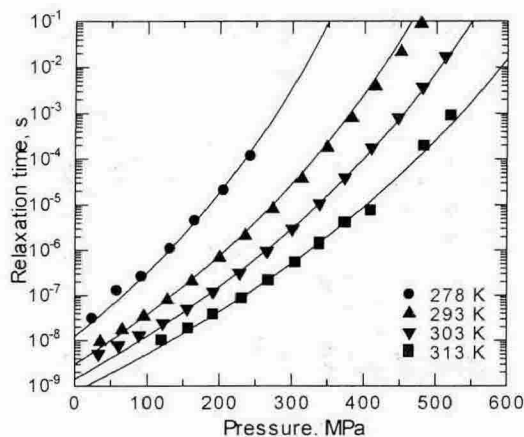


Fig. 2 Variation in dielectric relaxation time as a function of temperature and pressure for TCP.

### 3.3 Viscosity prediction from dielectric relaxation data

It is known that temperature and pressure dependence of the dielectric relaxation time is very similar to that of the viscoelastic relaxation time. Now let us suppose following relation is true.

$$\frac{\tau_{mech}(T, P)}{\tau_{mech}(T_r, P_r)} = \frac{\tau_{diel}(T, P)}{\tau_{diel}(T_r, P_r)} \quad (6)$$

where  $\tau_{mech}$  and  $\tau_{diel}$  are mechanical and dielectric relaxation time, respectively. Subscript  $r$  refers to the reference state. Since the viscoelastic relaxation time is ratio of zero frequency viscosity,  $\eta_0$ , and infinite frequency shear modulus,  $G_\infty$ , the zero frequency viscosity at temperature  $T$  and pressure  $P$  can be expressed by following formula:

$$\eta_0(T, P) = \eta_0(T_r, P_r) \frac{G_\infty(T, P)}{G_\infty(T_r, P_r)} \frac{\tau_{diel}(T, P)}{\tau_{diel}(T_r, P_r)} \quad (7)$$

For viscosity calculation, bulk modulus,  $K_0$ , had been used instead of  $G_\infty$ , because measurement of  $K_0$  is much easier than that of  $G_\infty$ . We assume that  $K_0$  and  $G_\infty$  change similarly with temperature and pressure as follows:

$$\frac{G_\infty(T, P)}{G_\infty(T_r, P_r)} = \frac{K_0(T, P)}{K_0(T_r, P_r)} \quad (8)$$

High-pressure viscosities were calculated using equations (7) and (8). Figure 3 shows the results of viscosity prediction. Plots in this figure show the viscosity measured using falling-sphere viscometer. The predicted results showed relatively good agreement with viscosity data. Dielectric relaxation is useful method for high-pressure viscosity estimation.

### REFERENCES

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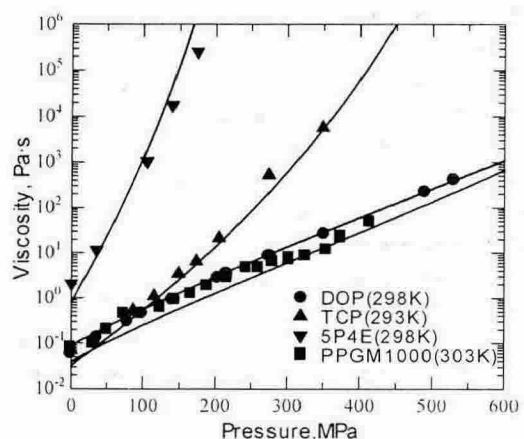


Fig. 3 High-pressure viscosity predicted from temperature and